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RADIOISOTOPE FUELED THERMOELECTRIC GENERATORS

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During the recent years a great attention has been paid to the design of low power self-contained sources utilizing the decay energy of radioisotopes . Since these power sources are extremely reliable and have a longer service life not requiring an additional charge they may be used to supply power to remote maintenance-free devices.

This report describes two radioisotope fueled thermoelectric generators designed in the USSR. The first power source utilizes Po-210 as a fuel, while the second one – Ce^{144}.

I. Radioisotope power source using Po^{210}

For recent years many papers have been published describing various radioisotope fueled thermoelectric generators delivering from 1 to 100 W. Lead selenides and tellurides are used as a thermoelectric material because of their higher figure of merit as compared with the well known materials. The figure of merit however is not the only factor indicative of thermoelectric material suitability. Application of high temperature alloys (although with the less figure of merit) allowed to increase the emitter temperature and so to save the weight of the device the efficiency being the same due to the greater temperature difference between hot and cold junctions.

A number of thermoelectric generators were designed with the initial power up to 10 W , they had similar thermoelectric conversion systems and differend only in a radiator and in the way the thermopiles are pressed to the hot surface. These instruments were used to determine equipment capacity, electrical parameters and temperature distribution through the blocks by means of a radioisotope heat source simulator.

Principle parameters. The preliminary investigation of the device versions have showed that a design where a flat capsule is snuggly fitted between hot surfaces of two thermopiles the cold junctions of which are attached to the radiator is a simplest one; this device has sufficiently low heat loss.

The capsule has the shape of rectangular parallelepiped of 60x60x13 mm. Manufacture technique and operating conditions limit to some extent possible values of its parameters. For example to make a capsule reliable at high pressure (up to 200 atmospheres) caused by helium produced in the decay its surface temperature should not exceed 850°C. Temperature-resistant rubber used for sealing the device limits the temperature of the radiator , which should not be more than 250°C.

A semiconductor section of the thermoelectric elements was chosen 3 cm long; if the length were less the power would fall off due to the higher commutation resistance in the total thermal battery resistance and the insulation plate resistance fraction in the thermopile resistance. On the other hand, if the length 1 is more than 3 cm the weight of the device builds up but its electric characteristics are improved slightly. Silicon-germanium alloy used in this device has an average figure of merit $Z = 4.10^{-4}$ $1/^{\circ}C$ and a thermal conductivity factor $x = 1.5.10^{-2}$ cal/cm.sec.°C.

The results calculations made it possible to choose the principle parameters, temperature characteristics and efficiency of the device . The results are listed in Table I.

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Conversion device. The silicon-germanium alloy was used as a thermoelectric material of the conversion device because of the following factors: a) high operation temperature (above 1000°C); b) low vapor pressure at T=1000°C; c) high strength; d) resistivity against radiation; e) low specific gravity. Each battery of thermogenerator comprises 16 thermoelements connected in series (8 thermoelements are of 'p'-type and the other eight of 'n'-type), they are screwed between two metal plates. The hot surface plate is made of carbon steel and the fixing screws are of molybdenum. The cold surface plate and its fixing screws are made of copper. Thermoelements screwed between steel and copper plates are coupled by molybdenum wire contact welded with the nickel foil 0.02 mm thick.

Both batteries are connected in series. In the installation 5 this connection is provided by a copper contact insulated with ceramics; in installations 6 and 7 the housing is used as a connecting conductor.

Installation housing. The housing of the installation 5 is of duraluminium and is cylindrical in shape, its bottom is provided with a flange to form sealing with a copper top of the housing. Two metal electrodes with ceramic insulation are welded into the copper top. The shape of the copper top flange is so chosen that the bottom can move 2-3 mm with respect to the flange due to the top deformation. Gas-tight coupling between the top and the housing is provided by a sealing ring of temperature-resistant rubber.

Since the housing of installations 6 and 7 are of duraluminium throughout, they are more rigid and lighter, the thermopiles are closer pressed to the radioisotope heat source capsule. The thermobatteries of the installation 5 are pressed to the radioisotope heat source capsule by means of a disk steel springs fitted on bolts coupling two parts of the housing. To provide good thermal contact between the elements of the installation 7 the both parts of the housing are coupled by a clamp secured to the flanges. The

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low section of the housing has stiffening ribs. The construction of the top section permits bending of the bottom with respect to the cylinder flange at a distance of 2 mm. The holes are provided in the housing to remove air from the interior and to fill it with the thermoinsulating material "Perlit". To increase blackness factor the housing is coated with a thin layer of hot proved paint with $\mathcal{E}$ =0.85. Fig.I shows general view of the installation 7.

Electrical Simulator Used for Capacity Tests of Generator. The electrical simulator of the radioisotope heat source is a graphite unit (size: 60 x 60 x 13 mm) with a molybdenum hairspring. Heater power was determined by connecting a a.c. wattmeter into the heater circuit. The performances of these systems were shown to be stable during 1000 hours operation. The results are listed in Table 2. The efficiency and electric power of thermogenerator 6 with the nuclide heat source simulator were also determined under various temperature conditions. Fig.2 shows the test results.

Radioisotope Heat Source. Radioisotope heat source is a rectangular parallelepiped; it is made of stainless steel. Capsules containing Po^{210} are inserted in five cells. The nickel capsule containing 1000-2000 curies of Po^{210} is mounted within two stainless steel tubes for strength. Tubes are provided with screw stoppers, welded during assembling. Fig.3 shows a general view of the radioisotope heat source and its components. The source thermal power was determined callorimetrically.

Tests of Thermoelectric Generator with Radioisotope Heat Source . Generator 7 has undergone tests. The thermogenerator with a nuclide heat source containing Po^{210} the original thermal power of which equaled 244 W was set up for 50 minutes, then it was connected to the meters. According to the bench test of generator 7, the following parameters wer continuously controlled: the voltage produced by the thermogenerator across the load 0.34 ohm; the hot skin temperature (two points); the radiating housing tempe-

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rature (two points); the gamma-radiation dose-rate at a distance of 0.3 m from the source. The test results are listed in Table 3.

Fig.4. presents volt-ampere characteristics taken periodically (in five-seven days).

II. Electric Power Generating System Containing Ce^{144}

At present time the human life is hardly possible without reliable and serviceable weather stations situated both in inhabited regions and in remote almost inaccessible ones, where there are no constant electric power sources. The available power sources have definite limitations hampering their usage and can not operate reliably and continuously.

Electric power nuclide sources based on the thermoelectric principle of converting the thermal power into electricity are more reliable and have sufficiently high electrical parameters.

The generator considered is a prototype of an electric power source for automatic meteorological stations. The possibilities of generators utilizing short lived beta-emitting nuclides were determined by a test model with a heat source containing Ce^{144} during controllable period (constant output power) and uncontrollable one (output power falls off).

Performances. By the beginning of an uncontrollable period the nuclide fueled generator contained 17500 curies of Ce^{144}. Fig.5 presents the generator general view.

During an uncontrollable operation electric output is 5.0-5.6 W; voltage - 3.5 V ; overall efficiency 3.5 - 4%. A thermopile based on solid d solutions of $Bi:Bi_2Te_3+Bi_2Se_3$ ('n'-type conductivity) and Bi_2Te_3 + Sb_2Te_3 ('p'-type conductivity) are used to convert the heat into electricity. A special automatic device provides a predetermined power level. The electric power is consumed by a storage battery with a capacity of 12 A/hours, this battery supplies power to a weather station, working in pulsed operation with 26 V and 6 A. 318

Radiation shielding consists of operating and shipping containers. The operating container shielding was designed to provide a dose rate not more than 1.0 r/hr at a distance of one meter from the generator surface. While the operating container inserted into the shipping one the dose rate reduces to the value not exceeding 10 mr/hr at a distance of one meter from the shipping container surface. The operating container has removable stiffening ribs, and the shipping one - fixed stiffening ribs. The generator may be used inserted into the operating container and into shipping one . Tungsten and lead are used as shielding material. The total weight of the generator with the operating container is 400 kg, the shipping container weight is 1200-1400 kg.

Main Parameters and Engineering Design . A necessary electric power of the generator is calculated assuming that the diurnal values of consumed energy and the delivered one are equal. The values of a necessary thermal power and an electric power for one disintegration assuming the gamma-absorption in the fuel, capsule and heat unit determine the radioisotope activity.

While constructing the device a particular attention has been payed to ensure a required radiation shielding, a minimum temperature drop between a cold junction and an ambient air, a lighter weight and smaller overall dimensions. Several designs were considered having various tungsten heat block-to-lead shielding thickness ratio. The constructions differed in the way the thermopiles are replaced, in the thermal regulation (radiation, thermal shunt, insulation thermal conductivity); in the shape and material of the radiation shielding (spherical and cylindrical geometry); lead, tungsten and depleted uranium were considered as shielding materials.

The generator comprises: a) a heat and a radioisotope blocks; b) a thermopile; c) a thermal power controller; d) an arrangement for substitution of the thermopile; e) housing; f) a biological shielding.

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A radioisotope block is fitted within a tungsten cylindrical heat block with a diameter of 100 mm, its sides are 20 mm and bottom is 40 mm thick. The cylinder is closed with a hemispherical top fixed to the cylinder with a covering ring.

A isotope block is a double capsule made of stainless steel XI8H9T-type. It consists of two coaxial cylinders within which a vessel is inserted filled with a melted cerium molybdate. Each cylinder is closed with a top, argon-arc welded and is checked for air-tight sealing. The heat block is mounted on the thermopile consisting of 97 thermoelements. The thermopile is fixed in a special case and attached to the bottom made of cylindrical copper plug. In order to maintain nominal temperature conditions the generator is provided with thermal control means including special thermal shields and a control actuator with a reducing gear. A constant heat flux through the thermopile (when isotope block power falls off) is assured by reducing a radiating surface of the heat block by moving the thermal shield. Fig.6 presents the thermal power vs time. The spring thermal control actuator is driven electrically. A thermal block, a thermopile and an arrangement for substitution this thermopile are inserted into a cylindrical housing with welded branches to mount a case with a thermopile. A cap with a lead filler closes the top end of the housing. Operation and shipping containers provide a radiation shielding. Stiffening ribs of aluminium alloy are used as a heat-removing system . This system consists of 64 rectangular ribs fixed with a coupling bolt on the operating container; these ribs provide a required temperature drop between an ambient air and a thermopile base. The conjugate surfaces of the dismountable elements are highly finished and a sufficient specific pressure is ensured at the point of contact.

<u>Thermophysical Calculations</u>. Cylindrical heat source temperatures were calculated from a thermal conductivity differential equation. Heat flux distribution over generator blocks were also determined. This distribution depends upon the thermopile operating conditions i.e. free running, rated operation, and short circuit.

The results are listed in Table 4.

4. Heat Generation Calculation

The heat generation in the unit, containing Ce^{144} is determined by beta-particles of $Ce^{144} + Pr^{144}$ and gamma-rays, absorbed in both isotope and heat blocks.

Space distribution of the specific heat generation due to beta-emission may be considered uniform throughout the capsule interior. Heat leakage due to bremsstrahlung are neglected since a bremsstrahlung transfered energy equals 1–2 per cent of the total energy; the most portion of this energy is absorbed in isotope and heat blocks. Space distribution of the specific heat generation due to gamma-radiation (it is of particular importance for nuclides of the Ce^{137} type) is defined by the following expression:

for gamma-radiation absorbed by the capsule material

$$Q_{\gamma 1}(\vec{r}) = \frac{3{,}7\cdot 10^{10}}{4\pi} kq \sum_E E n(E) \mu_a(E) \int_V \frac{e^{-\mu(E)|\vec{r}-\vec{r}'|}}{(\vec{r}-\vec{r}')^2} B_a\left[\mu(E)\left|\vec{r}-\vec{r}'\right|\right] d\vec{r}'$$

Here $Q_\gamma(\vec{r})$ - heat generation per a volume unit close to the point with coordinate $\vec{r}$ (W/cm^2); $k = 1.6 . 10^{-13}$ W.sec/Mev; q - material specific activity, curies/cm^3; n(E) - number of gamma-rays per one disintegration; V - capsule core volume; E - gamma-ray energy; μ (E) and μ_a (E) - gamma-ray attenuation coefficient and energy-absorption coefficient in the fuel respectively; $B_a[\mu(E)\, / \vec{r} - \vec{r}'\, /]$ accummulated absorbed energy factor for gamma-radiation in the fuel.

Similar relations exist for gamma-rays absorbed by the capsule walls $Q_{\gamma 2}(\vec{r})$ and by the heat block $Q_{\gamma 3}(\vec{r})$.

The total heat generation is calculated by integrating the expressions for $Q_{\gamma 1}(\vec{r})$, $Q_{\gamma 2}(\vec{r})$ and ;$Q_{\gamma 3}(\vec{r})$ over drug, capsule and heat block volumes respectively. The heat generation during rated operation (controllable period end) was 135 W (17500 curies).

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Thermal power of the isotope block was measured by a special calorimeter, the obtained data agreed with the calculated ones.

<u>Radiation Shielding Design</u>. A multigroup method was used to design shielding, partial gamma-ray constants (the dose-rate given by the separate gamma line of the unshielded isotope) corresponding to gamma lines of $Ce^{144}+Pr^{144}$ was first determined. Bremsstrahlung was taken into account by replacing a continuous spectrum with a discrete one for each line of which a corresponding gamma-constant was determined. A series of partial gamma constants obtained was treated according to "competing lines" method that resulted in time saving.

The necessary heterogeneous shielding thickness was calculated from

$$\frac{[P]}{k} = q \sum k_{\gamma i} \; L_i \, B_i \,, \qquad (I)$$

where P - permissible dose rate; k - assumed safety factor ; q - drug specific activity ; k - partial gamma-constant; L_i - radiation function of the cylindrical source behind the shielding allowing for geometric factor, self-absorption in the source and attenuation in the shielding disregarding the scattered radiation (radiation function was calculated by an electronic computer); B_i - dose buildup factor allowing for multiple scattering of gamma-radiation within both the drug and capsule substance and the shielding.

The following expression was used for a dose buildup factor in a heterogeneous medium

$$B_i = \sum_{n=1}^{N} B_n \left(\sum_{k-1}^{n} \mu_{ik} d_k \right) - \sum_{n=2}^{N} B_n \left(\sum_{k=1}^{n-1} \mu_{ik} d_k \right), \qquad (2)$$

where N - number of heterogenous shielding layers; μ_{ik} - attenuation coefficient of K-layer; d_k - K-layer thickness.

Expressions (I) and (2) were used to determine a dose rate on the shielding surface. It is necessary to notice that thicknesses calculated by this way agree with thickness values estimated according to Monte-Carlo method.

Table 5 lists calculated values of two different shielding.

Thermoelectric generator and Voltage Transformer. A thermoelectric conversion device consists of 97 thermoelements (5 mm x 5 mm) , 20 mm high. The thermoelements are made up of solid solutions based on triple alloys $Sb_2Te_3+Bi_2Te_3$ and $Bi_2Te_3 + Bi_2Se_3$. A solder provides commutation allowing to pperate in the temperature range of 300°C. To prevent the thermoelements from oxidation at high temperatures hot junction are coated with a thin layer of a special enamel. The total heat losses of the generator were calculated allowing for the heat losses across the thermopile insulation and the other blocks. The generator tests carried out by means of an electrical simulator of the heat source under normal operation conditions showed good agreement between calculated and experimental data. Fig.7 presents generator load characteristics obtained at controllable operation, fug.8 - the same ones during the uncontrollable period after 50 days of operation under these conditions.

To recieve a d.c. voltage of 26 V this system comprises a transistorized dc-dc transformer. Energy storage is accomplished by silver-zincum accumulators. The transformer supplies current to the storage battery depending on the operating conditions of the meteostation. The transformer efficiency is 75 per cent; the accumulator efficiency is 75-80 per cent. The higher overall efficiency of 98 per cent achieved with accumulators connected in series .

Operation. The above generator with accumulators and a voltage transformer is used as a power source at an automatic weather station located in the middle part of the Soviet Union. To minimuze a dose rate around the generator it is slightly buried into the ground. The generator operation was controlled by the output instruments which indicated the principle parameters: voltage, electromotive force, current All output parameters of the dc-dc transformer and accumulators were automatically recorded.The operation recured in every two hours. The output power at the end of controllable period was 5.4 W , voltage was 3.6 V.

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Fig.9 shows variations of the dc-dc transformer input voltage (9-a), of the accumulator battery output voltage (9-b) and of the transformer efficiency for the whole cycle of operation . During an uncontrollable operation after two month period of 2 hr cycle the meteo station was transffered to 3 hr cycle. Climatic conditions were controlled during this experimental operation. The heat removal depended on these conditions. At present the system is in operation and if the power falls off the intervals between the operation should be made longer.

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List of References

1. Barnat M. et al. Nucleonics, 17, No.5, 166 (1959).
2. Harvey D.G., Horse I.G., Nucleonics, 19, No.4, 62 (1961).
3. Harvey D.G., Carpenter R.T., IAS Paper, No.61, 18(1962).
4. Иоффе А.Ф. Полупроводниковые термоэлементы. Изд.АН СССР, 1956.
5. Джелепов Б.С., Пекер Л.К. Схемы распада радиоактивных ядер, Изд-во АН СССР, 1958.
6. Фрадкин Г.М., Кулиш Е.Е. Источники α, β, γ и нейтронных излучений, Госатомиздат, 1961.
7. Geiger I.S. et al, Nuclear Physics, 28, 387-406 (1961).
8. К.К.Аглинцев, В.М.Кодюков, А.Ф.Лызлов, Ю.В.Сивинцев. Прикладная дозиметрия, Госатомиздат, 1962.
9. Кутателадзе С.С. Основы теории теплообмена, Машгиз, 1962.
10. Бурштейн А.И. Физические основы расчет полупроводниковых термоэлектрических устройств, Физматгиз, 1962.
11. Рогинский В.Ю. Электропитание радиоустройств, Связьиздат, 1963.
12. Collin W., Physics Letters, 5, 329 (1963).
13. Гусев Н.Г., Ковалев Е.Е. и др. Защита от излучения протяженных источников. Государственное издательство литературы в области атомной науки и техники, 1961.

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Table I

1. Insulation thermal resistance, $\frac{^{o}C.sec}{cal}$	12
2. Heat flow through insulation, W	49
3. Initial heat power of the capsule, W	320
4. Radiator surface, cm^2	900
5. Number of thermoelements (allowing for the external load voltage of 1.8 V)	32
6. Thermopile cross section, cm^2	0.8
7. Hot junction temperature, ^{o}C	760-775
8. Cold junction temperature, ^{o}C	300-330
9. Thermoelectric generator efficiency, %	3.5-3.9
10. Overall efficiency, %	3-3.3

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Table II

Nos.	Parameters	Installation number		
		5	6	7
1	2	3	4	5
1.	Dimensions, mm			
	height	128	126	210
	diameter	195	196	190
2.	Weight (without the radioisotope heat source capsule), kg	3.1	2.7	2.8
3.	Simulator heat power, Q, w			
	a) heat power equivalent of the radioisotope heat source containing 10000 curies of Po^{210}	320	320	320
	b) heat power equivalent after three month of operation	205	205	205
4.	Electrical power of the installation, W:			
	a) when Q = 320 W	10.15	10.9	9.65
	b) when Q = 205 W	4.35	4.67	4.15
5.	Efficiency when Q=320 W,%			
	a) thermoelectric	3.75	4	3.56
	b) thermal	85	85	85
	c) overall	3.18	3.41	3.02
6.	Electromotive force when Q = 320 W, V	3.9	3.6	3.8
7.	Electromotive force when Q = 205 W, V	2.42	2.24	2.36
8.	Maximum temperature of the thermoelements hot plate when Q =320 W, ^{o}C	861	840	817

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1	2	3	4	5
9.	Radiator maximum temperature when Q = 320 W, °C	241	250	240
10.	Maximum temperature of the thermoelement hot plate when Q ≅ 205 W, °C	670	650	633
11.	Radiator maximum temperature when Q = 205 W ,°C	190	196	195

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Table III

Nos.	Parameters	Operation period, hours			
		48	1000	1400	2000
1	Thermal power of the radioisotope heat source, W.	244	200	185	163
2	Electromotive force, V	2.85	2.3	2.09	1.88
3	Load voltage 0.34 ohms, V	1.4	1.14	1.04	0.92
4.	Power output, W	5.8	3.79	3.24	2.52
5	Installation efficiency, %	2.36	1.91	1.75	1.55
6	Thermoelectric generator hot plates temperature when operating at the load of 0.34 ohms,	759	633	600	545
	^{o}C	746	542	503	440
7	Radiator housing temperature, ^{o}C	230	202	192	163
		216	190	178	157
8	Activity of the radioisotope heat source using Po^{210}, curies	7700	6300	5320	5140

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Table IV

Operating conditions	Thermopile temperature drop	Heat flux via thermopile, W	Heat flux via the other blocks, W	Thermal efficiency
Free running	225	112.5	22.5	83.4
Rated operation	190	119	16	88
Short circuit	170	121	14	89.6

Note: Cold junction temperature assumed to be 0°C, the ambient temperature - +20°C. Electric power of the thermopile is of 5.2 watts.

Table V

Direction	Version	Container type: Operating	Container type: Shipping
Axial	1	Pb - 12 cm	Pb - 9.8 cm
	2	W-1 cm + Pb-10.I cm	Pb - 9.9 cm
Radial	1	Pb - 11.8 cm	Pb - 9.8 cm
	2	W-2 cm + Pb - 8.2 cm	Pb-9.9 cm

Note : Ce^{144} total activity of the source is 30000 curies; a dose rate is 1 r/h and 10 mr/h at a distance of 1 meter from the operating container and the shipping one respectively.

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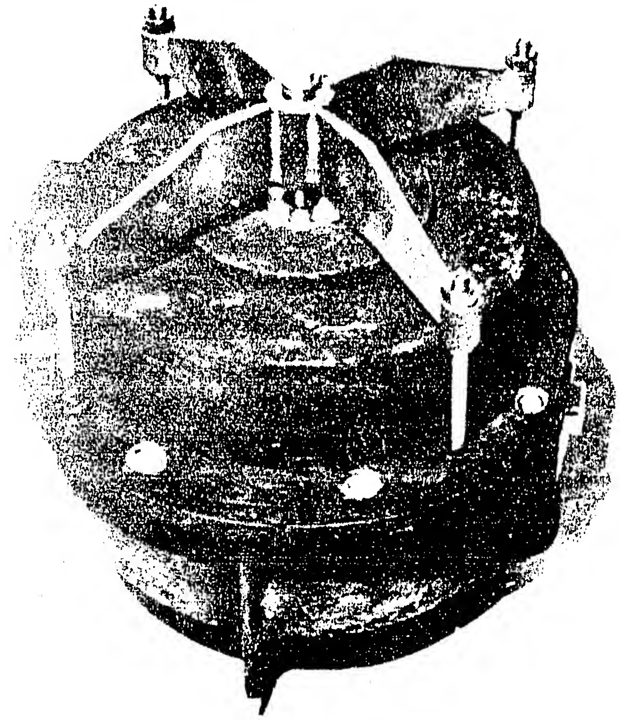

Fig.1. General view of the generator, containing Po^{210}

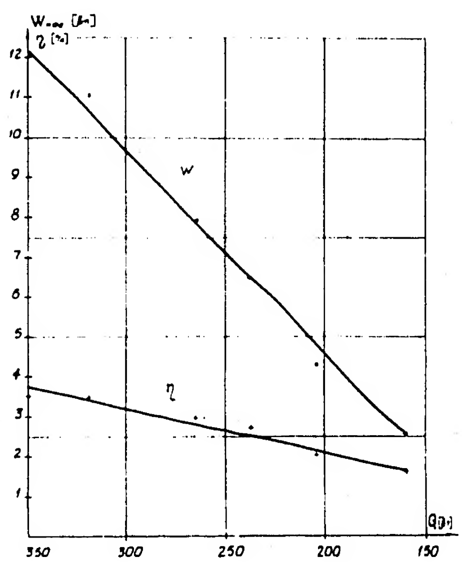

Fig.2. Electric power and efficiency of the installation under various thermal conditions

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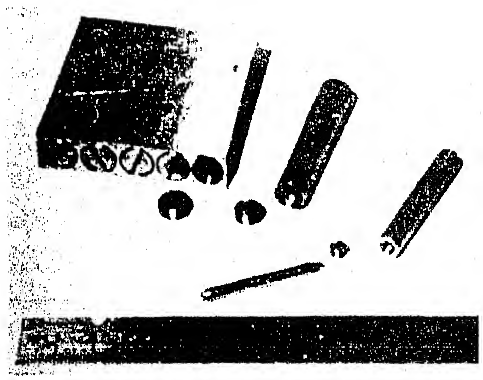

Fig.3. Heat source capsule and its components

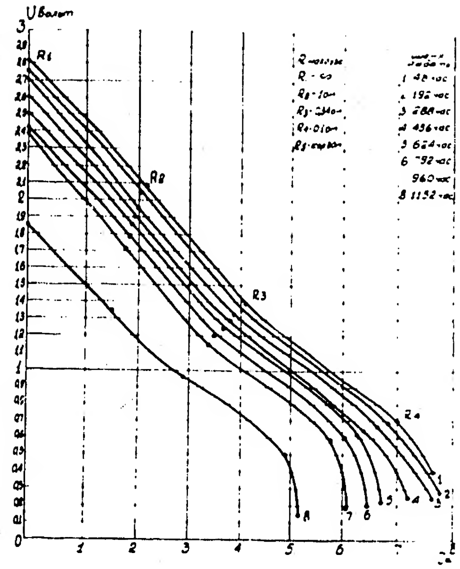

Fig.4. Volt-ampere characteristics

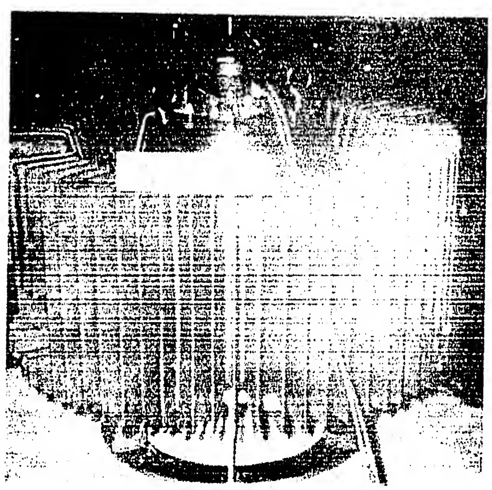

Fig.5. Generator general view using Ce^{144}

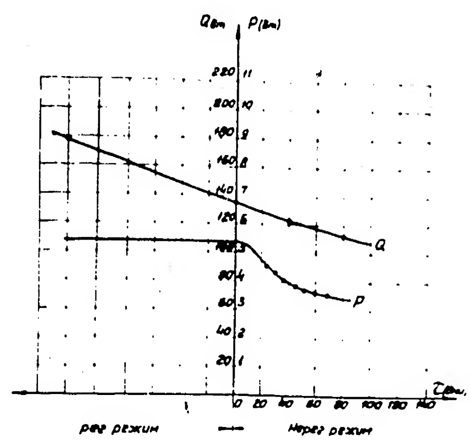

Fig.6. Thermal (Q) and output electric (P) power v.s. operation time

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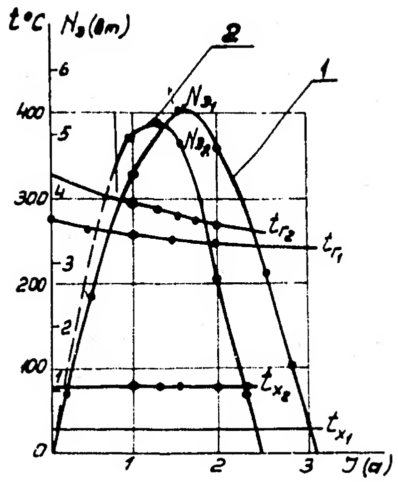

Fig. 7. Generator load characteristic under controllable conditions
1 - ambient air temperature +10^{o}C
2 - ambient air temperature +60^{o}C

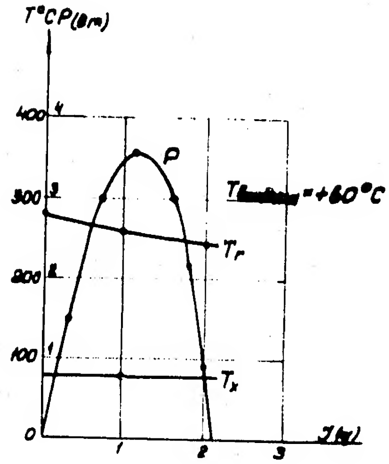

Fig.8 Generator load characteristic under uncontrollable conditions

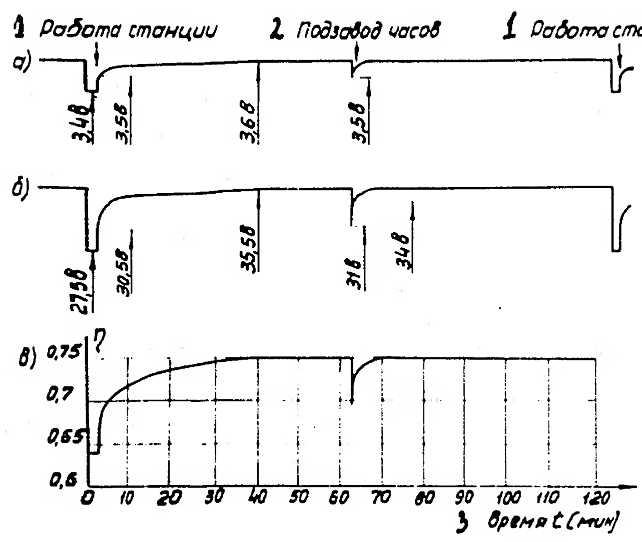

Fig.9. The pattern of change of DO-DC transformer input voltage(a), accumulator battery output voltage(b) and transformer efficiency (c) for one operating cycle of the weather station
1 - station operation; 2 - clock winding up
3 - time(min)